

- [54] CORROSION RESISTANT ALLOY
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[56] **References Cited**
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[57] **ABSTRACT**

A corrosion resistant alloy is provided having as essential elements about 0.8 to 1.2% carbon, 1 to 2.2% silicon, 0.4 to 10% manganese, 23 to 35% chromium, 2.5 to 11% tungsten, 1 to 4% molybdenum and the balance essentially iron. Other elements may be optionally present, such as up to about 2% each of cobalt, nickel, niobium, tantalum, vanadium, titanium, up to about 1% aluminum, up to about 0.1% boron, up to about 1% nitrogen, up to about 4% copper, rhenium in amounts of up to about 30% of the tungsten content, and up to about 3% of misch metal.

9 Claims, No Drawings

CORROSION RESISTANT ALLOY

This application is a continuation-in-part of Ser. No. 186,476, filed Oct. 4, 1971, now abandoned.

This invention relates to a corrosion resistant alloy having particular use in atmosphere containing one or more of vanadium, sulfur and sodium, such as occur in the combustion of certain fuel oils.

State of the Art

It is known that the combustion of heavy oil containing vanadium, sulfur and sodium present corrosion problems with regard to metals in contact with the combustion products thereof. As such oils are cheap and economically advantageous, it would be desirable to provide alloys having improved resistance to corrosion in such atmospheres, especially for use as exhaust valves in diesel engines or structural elements, e.g. furnace parts, which are used in contact with such corrosive oils.

Corrosion resistant alloy materials have been proposed for valves for use in internal combustion engines. Generally, such materials have been of the iron-base austenitic variety, the alloys containing chromium, manganese and carbon. Normally, the alloys also contain silicon and nitrogen. Thus, in U.S. Pat. No. 3,149,965, an austenitic valve steel is disclosed consisting essentially of about 0.45% to 1.5% carbon, about 12% to 30% chromium, about 5% to 20% manganese, about 3.25% to 6% nickel, about 0.1 to 0.6% nitrogen, about 0.5% to 1.5% silicon, and the balance substantially all iron, the various ingredients being proportional to assure a substantially fully austenitic structure. As regards other austenitic steels which have been proposed, reference is made to U.S. Pat. Nos. 3,165,401 and 3,340,046. Additional patents disclosing heat and wear resistant iron-base alloys are U.S. Pat. Nos. 1,460,048 and 1,671,417.

While many attempts have been made to provide iron-base alloys, especially for use in diesel engines, they have not been wholly satisfactory in providing the desired corrosion resistance for exhaust valves of medium-sized diesel engines in which exhaust gas temperatures are normally above 500°C. Generally, inferior results are obtained when such engines employ cheap, heavy fuel oil.

The vanadium content of such heavy oils having a viscosity of at least or more than about 1,500 Redwood seconds ranges from about 50 to 1,000 ppm (parts per million), and usually from about 50 to 100 ppm. While the amount of corrosion appears to be directly proportional to the vanadium content, the amount of corrosion is generally considerable even when very small amounts of vanadium are present in the fuel oil at the low end of the range stated hereinabove.

According to the present invention, markedly improved resistance to corrosion has been obtained in combustion atmospheres containing vanadium, sulfur and sodium compared to previously known alloys, even in the presence of high gas velocities and under aggravated conditions of gas erosion.

It has been found that by providing an iron-base alloy characterized by the presence of ferrite and sigma phases, instead of austenite, relatively high hardness is assured. It has generally been the practice to avoid the formation of ferrite and sigma phases and to increase the hardness of prior alloys by heat treatment. How-

ever, this is not necessary with the alloy of the invention as it has relatively high intrinsic hardness in the non-austenitic state.

Objects of the Invention

It is thus the object of the invention to provide a corrosion resistant, non-austenitic iron-base alloy characterized by the presence of ferrite and sigma phases and also characterized by particular resistance to combustion atmospheres containing one or more of the corrosive materials vanadium, sulfur and sodium.

Another object is to provide a non-austenitic, iron-base corrosion resistant alloy for use on at least the working face portions of exhaust valves in internal combustion engines, such as diesel engines.

A further object is to provide as an article of manufacture a structural element, such as furnace elements or parts, for use in corrosive environments containing such corrosive agents as vanadium, sulfur, sodium and the like.

A still further object is to provide an exhaust valve, at least the valve seat portion of which is characterized by an alloy composition having improved resistance to corrosion in engines using fuel oil containing such corrosive agents as vanadium, sulfur and sodium.

These and other objects will more clearly appear when taken in the light of the following disclosure and the appended claims.

Statement of the Invention

Stating it broadly, the novel non-austenitic alloy provided by the invention comprises as essential alloying elements by weight about 0.8 to 1.2% carbon, about 1 to 2.2% silicon, about 0.4 to 10% manganese, about 23 to 35% chromium, about 2.5 to 11% tungsten, about 1 to 4% molybdenum, and the balance essentially iron. Other alloying elements may be optionally present, such as up to about 2% each of the elements selected from the group consisting of cobalt, nickel, niobium, tantalum, vanadium and titanium and in addition up to about 1% nitrogen, up to about 4% copper, rhenium in amounts up to about 30% of tungsten present in the alloy and up to about 3% total of misch metals. Thus, rhenium may range up to a maximum of about 0.3 × 11% W or up to about 3.3%.

Laboratory tests of exhaust valves produced in accordance with the invention in actual test engines have shown the alloy of the invention to be markedly superior to nickel and cobalt-base alloys (e.g. stellites). The latter alloys were not suitable for use in exhaust valves.

The range of the chromium content of 23 to 35% is important in the present alloy. Chromium substantially below 23% results in inferior corrosion properties and hardness, while amounts substantially over 35% tend to cause brittleness. The range of 1 to 4% molybdenum and tungsten from about 2.5 to 11% provides good high strength properties and good resistance to gas erosion. The absence of these elements results in inferior high temperature strength and inferior resistance to erosion. Substantial amounts of molybdenum may adversely affect the corrosion resistance, while excessive amounts of tungsten may lead to welding problems.

As will be noted, the carbon content is relatively high and is controlled over the range of 0.8 to 1.2% by weight. The carbon content is important in that substantially below 0.8%, insufficient carbides are formed, thus resulting in inferior hardness. Excessive amounts

of carbon substantially above the maximum tend to embrittle the alloy and also adversely affect its weldability. In addition, excess carbon results in the carbides being easily broken or dislodged when the part is subjected to mechanical stress and erosion.

Silicon in amounts substantially below the minimum results in lower hardness and less resistance to corrosion. Too much silicon adversely affects weldability. The manganese content over the aforementioned range of 0.4 to 10% beneficially affects the hardness and corrosion resistance of the alloy.

The amount of nickel and cobalt, if present, is limited to a maximum of 2% each; otherwise, excessive amounts of these elements have an unfavorable effect on the corrosion properties.

Niobium, tantalum, vanadium, and titanium in amounts up to about 2% each act as carbide formers. The presence of boron and nitrogen in small amounts stated in the ranges disclosed hereinbefore will increase the hardness of the alloy. Copper should not exceed 4% by weight as too much copper adversely affects the corrosion resistance. Aluminum should preferably not exceed 1%.

It has been unexpectedly found that a particularly fine grained structure can be obtained in the alloy by adding an amount of rhenium equivalent to about 3% of the tungsten content. While it is not clear how the rhenium influences the alloy, an improvement in the resistance to corrosion and gas erosion is also obtained. The rhenium may be added in amounts up to about 30% of the tungsten content, e.g. up to 3.3% by weight, without appreciably changing the beneficial effects thereof.

Beneficial results have also been obtained by the addition of misch metal in an amount of up to about 3% by weight. An example of misch metal is one containing 48.1% cerium and 51.9% of the rare earth metals, such as lanthanum and the like. The presence of misch metal also results in a fine grain with a resultant increase in hardness, the beneficial effects being improved resistance to gas corrosion and erosion.

DETAILS OF THE INVENTION

The alloy of the invention has been tested in comparison to a great number of other alloys, particularly commercial alloys. These tests have included welding tests, structure determination, hardness measurements and corrosion measurements, including a relative estimate of the life of the alloy under corrosive conditions. The life test is carried out relative to the life of an alloy referred to in the trade as Stellite 20 (2.5% C, 33% Cr, 18% W and 46.5% Co), an alloy previously known for use in valves. In order to obtain a relative comparison, the life of Stellite 20 is set at 10.

The laboratory test comprises testing a weld deposit of the alloy in a melt formed of V_2O_5 and Na_2SO_4 1 to 1 weight ratio) maintained at 700°C. The test is carried out for 336 hours, the alloy test piece removed, cleaned and the weight loss determined. The effects of corrosion were studied using a metallographic microscope, a microprobe device and a scanning electron microscope. The hardness was measured before and after the corrosion test. However, in most cases, no change in hardness was noted.

The results of the laboratory tests are summarized in Tables 1A and 1B which follow, Table 1A listing the compositions of alloys within the invention (Nos. 1 to 9) and alloys outside the invention (Alloys A to Y), while Table 1B summarizes the results of the tests. Alloy B represents Stellite 20, its composition being given as 2.5% carbon, 33% chromium, 18% tungsten and 46.5% cobalt. It will be noted from Table 1B that its relative life in the salt bath of $V_2O_5 - Na_2SO_4$ is given as 10. As will be noted, Alloys 1 to 9 fall within the range by weight of about 0.8 to 1% C, about 1.5 to 2% Si, about 1 to 10% Mn, about 23 to 32% Cr, about 1 to 3% Mo, about 2.5 to 9% W and the balance essentially iron, optional elements, such as Re, Cu, N, B, misch metal and V may be present as shown. A preferred alloy is one containing about 1.8 to 2.2% Si, about 0.4 to 10% Mn, about 28 to 35% Cr, about 7 to 11% W, about 1 to 4% Mo and the balance essentially iron.

TABLE 1A

ALLOYS OF THE INVENTION										
% ELEMENT										
LLOY	C	Si	Mn	Cr	Mo	W	Fe	Ni	Co	OTHERS
1	1	2	1	32	3	9	bal.	—	—	—
2	1	2	1	32	3	8.73	—	—	—	0.27 Re
3	1	2	1	32	3	9	bal.	—	—	2 Cu
4	1	2	10	32	3	9	bal.	—	—	—
5	1	2	10	32	3	9	bal.	—	—	0.9 N
6	1	2	10	32	3	9	bal.	—	—	0.1 B
7	1	2	1	32	3	9	bal.	—	—	3 misch metal
8	1	2	1	32	3	9	bal.	—	—	2 V
9	0.8	1.5	1	23	1	2.5	bal.	—	—	—
ALLOYS OUTSIDE THE INVENTION										
A	1	—	—	26	—	5	—	—	66	bal. residuals
B	2.5	—	—	33	—	18	—	—	46.5	—
C	0.25	—	1	29	5.5	—	—	—	62.5	2 Cu
D	2.8	1	1	27.4	—	—	—	0.2	61.5	2 Cu, 4.1 V
E	2.8	1	1	27.4	4	—	—	0.2	57.5	2 Cu, 4.1 V
F	2.3	0.5	0.5	32	4	12	—	—	45.7	3 Cu
G	2.3	0.5	0.5	32	4	11.6	—	—	45.7	3 Cu, 0.4 Re
H	2.8	1	1	27.4	—	—	—	0.2	61.5	2 Cu, 4.1 V
I	2	1	—	30	4	12	—	—	45	2 Cu, 4 V
J	1	2	—	30	4	13	—	—	48	2 Cu
K	1	1	—	30	5	13	—	—	48.1	1.9 Cu
L	1	1	—	30	5	12.6	—	—	48	2 Cu, 0.4 V
M	2.8	1	1	27	—	—	bal.	0.2	—	4.1 V
N	3	1	0.5	30	3	9	bal.	—	—	3 V

TABLE 1A - Continued

ALLOYS OF THE INVENTION										
% ELEMENT										
LLOY	C	Si	Mn	Cr	Mo	W	Fe	Ni	Co	OTHERS
O	3	1	1	30	3	9	bal.	—	—	3 V
P	3	1.15	1.02	30.4	2.8	9.4	bal.	—	7.1	3.5 V
Q	—	—	2	3C	1	—	bal.	10	—	—
R	3.2	—	—	29	—	—	bal.	—	—	—
S	2	—	—	30	—	—	bal.	4	—	—
T	3	2	1	32	3	9	bal.	—	—	—
U	0.5	2	1	32	—	5	39.5	20	—	—
V	1	2	1	32	12	—	52	—	—	—
W	0.1	—	—	17	17	5	6	55	—	—
X	1	4.5	—	15	—	—	7.5	bal.	—	3 B
Y	—	—	—	17	18	5	7	53	—	—

TABLE 1B

ALLOY No.	TYPE	H_v^{33} Kp/mm ² *	RELATIVE LIFE
1	Iron-base	460	28
2	"	510	31
3	"	490	26
4	"	510	33
5	"	550	27
6	"	530	28
7	"	480	31
8	"	470	25
9	"	320	24
OUTSIDE THE INVENTION			
A	Co-base	410	7
B	"	620	10
C	"	390	<10
D	"	510	"
E	"	610	"
F	"	630	"
G	"	660	"
H	"	450	"
I	"	500	"
J	"	620	"
K	"	420	"
L	"	520	"
M	Iron-base	480	20-22
N	"	580	"
O	"	440	"
P	"	400	18-19
Q	"	270	"
R	"	450	"
S	"	315	"
T	"	530	21
U	"	390	14
V	"	450	18
W	Ni-base	300	8-9
X	"	210	"
Y	"	290	"

*Vickers hardness in Kilopond per square millimeter

It will be noted from Table 1B that the relative length of the life of the alloys in the $V_2O_5 - Na_2SO_4$ melt is best for the iron-base alloys, whereas the cobalt and nickel-base alloys only attained one-third to one-half of the life of the alloys of the invention. Thus, the alloys within the invention (Nos. 1 to 9) exhibited a life relative to Stellite 20 (Alloy B - relative life of 10) ranging anywhere from 24 to 33.

Cobalt-base alloys A to L exhibited a relative life of up to 10, while the nickel-base alloys W to Y exhibited a relative life of 8 to 9.

Iron-base alloys M to V (outside the invention) exhibited a relative life of 8 to 22, with the majority of these alloys exhibiting relative life below 19. It will be noted that the addition of excess amounts of nickel and/or cobalt to these alloys lowers the relative life

(note P, Q, and U). Carbon in excess of 2% (e.g. 3%) has a deteriorating effect.

It will be noted from the alloys of the invention (Nos. 1-9) that a reduction of chromium to 23% (Alloy No. 9) gives acceptable corrosion resistance. Good results were obtained with an alloy containing 0.8% C, 1.5% Si, 1% Mn, 23% Cr, 4% W, 1% Mo and the balance essentially iron.

While 3% carbon in the iron-base alloy might give a relative life of 20 (double that of Stellite 20), mechanical tests show that the high carbon content is disadvantageous and, therefore, not of practical interest.

It appears from Tables 1A and 1B that a preferred alloy is one containing about 1% carbon, about 2% silicon, about 1 to 10% manganese, about 32% chromium, about 3% molybdenum, about 9% tungsten and the balance essentially iron. The foregoing alloy may contain optionally up to about 0.1% boron, up to about 1% nitrogen, rhenium in amounts up to about 30% of the tungsten content, up to 2% vanadium and up to about 3% misch metal.

Tests have indicated that small amounts of Al, Ti, Nb and Ta can be added without the properties of the alloy being adversely affected to any degree.

In addition to the foregoing, tests of exhaust valves utilizing the alloy within and outside the invention were carried in Diesel engines of the type S.E.M.T. Pielstick PC-2. The tests summarized in Table 2 were carried out in a land-based engine operating with heavy oil containing among other things 300 ppm vanadium, 100 ppm sodium and 3.2% sulfur. The engine was operated at full capacity whereby exhaust temperatures of more than 500°C were obtained for intervals of 150 hours, after which the exhaust valves were checked. As is the custom, some valves have hollow stems to enable them to be cooled during use.

The valves utilizing the alloy of the invention were not cooled and, in addition, were rotated about their axis while in use by means of a device. Duplicate tests were carried out in all instances.

Some of the tests were also carried out aboard ship. These tests were carried out in such a manner that 30 valves (uncooled) produced according to the invention were checked every 800 hours and the results compared to cooled standard valve surface-coated with a weld of Stellite 20 (note Table 3). The 30 valves of the invention (which were also uncooled) were rotated during use. The corrosion effects were observed and

correlated with the laboratory results. The physical strength properties were noted and the resistance to gas erosion observed.

The results of the land-based tests and the tests on board ship are given in Tables 2 and 3, respectively, as follows:

TABLE 2

% ELEMENT	CONVENTIONAL ALLOY		OUTSIDE INVENTION	THE INVENTION	
	AA	BB	CC	11	12
C	2.5	0.2	3	1	1
Si	—	—	1	2	2
Mn	—	—	1	1	1
Cr	33	30	30	32	32
Mo	—	6	3	3	3
W	18	—	9	9	8.73
Fe	—	—	bal.	bal.	bal.
Co	46.5	63.8	—	—	—
Ni	—	—	—	—	—
Others	—	—	3 V	—	0.27 Re
RE-SULTS					
After 150 hrs.	Obvious Corrosion	Heavy Corrosion	Material deformed and carbide grains dislodged. Not corroded	No Corrosion	No Corrosion
After 300 hrs.	"	"	"	"	"
After 450 hrs.	—	—	—	—	—

TABLE 3

TEST OF EXHAUST VALVES ON BOARD SHIP				
ALLOY COMP.	COOLED CONVENTIONAL ALLOY	UNCOOLED ALLOY OF THE INVENTION		
	2.5% C 33% Cr 18% W bal. Co	1% C 2% Si 1% Mn 32% Cr 3% Mo 9% W bal. Fe	1% C 2% Si 1% Mn 32% Cr 3% Mo 8.73% W 0.27% Re bal. Fe	
After 800 hours	Occasional valves corroded. (1-2 out of 10)	No Corrosion	No Corrosion	
After 1600 hours	Several valves corroded. Often totally replaced.	"	"	
After 2400 hours	Most of the valves replaced.	"	"	

It will be clearly apparent from Table 2 that the exhaust valves produced in accordance with the invention exhibit a marked improvement in resistance to corrosion as compared to the standard valve having a welded coating of Stellite 20 (Alloy AA of Table 2 as compared to Nos. 11 and 12 of the invention). As will be noted, Alloy AA showed obvious corrosion after 150 hours of testing, while alloys 11 and 12 of the invention showed no corrosion after 450 hours of testing, over three times longer in time and still no corrosion.

Alloy BB, also a cobalt-base alloy, exhibited heavy corrosion after 150 hours. The iron-base Alloy CC, which is similar to Nos. 11 and 12 except for the high carbon (3%) and 3% V exhibited poor physical properties in that the valve deformed and in that the carbide

grains were dislodged or torn loose due to applied mechanical stresses during use. Observations in the laboratory test valves showed a clear connection between the carbon content and microbrittleness.

Referring now to Table 3, it will be noted that valves using the alloy of the invention tested on board ship exhibited markedly improved performance compared to the standard valves coated with Stellite 20. Because these valves tend to have inferior resistance to corrosion, they have to be water cooled. Cooled valves are expensive to manufacture and also require careful attention during use, as the hose connections to the hollow stem must be checked and often replaced.

As will be clearly apparent from Table 3, the uncooled valves of the invention showed no corrosion after 2,400 hours of service. As regards the cooled conventional valve (Stellite 20), an occasional valve corroded after 800 hours; several valves corroded after 1,600 hours and often totally replaced, and, after 2,400 hours, most of the valves were replaced. Normally, in practice, a water-cooled valve is replaced or repaired after about 1,500 hours of operation. It is abundantly clear that the valve utilizing the alloy of the invention is markedly superior to the conventional alloy.

The preferred composition containing about 0.8 to 1.2% carbon, about 1.8 to 2.2% silicon, about 0.4 to 10% manganese, about 28 to 35% chromium, about 7 to 11% tungsten, about 1 to 4% molybdenum, and the balance essentially iron is particularly interesting. The foregoing composition and the other composition ranges stated herein, with or without the optional ingredients, are useful in producing structural elements subjected in use to the corrosive effects of combustion gases containing vanadium and optionally such corrosive ingredients as sulfur and sodium.

In protecting exhaust valves against corrosion and erosion, the alloy of the invention is weld-deposited upon the working portion of the valve head, that is, at least on the valve seat. The valve head is provided with an overlay of the alloy using the hard facing technique normally employed for that purpose, the alloy being weld deposited using a welding rod of the alloy. Thereafter, the weld deposit is finished by grinding.

Thus, one embodiment of the invention resides in an article of manufacture comprising an exhaust valve, at least the head of which above the stem at the valve seat portion is characterized by an overlay of the aforementioned alloy of the invention.

Another embodiment of the invention resides in a heat resistant structural element, for example, a furnace element, such as a support for a belt in a sintering furnace used for sintering pelletized iron powder or iron oxide ore, particularly in which the heat in the furnace is supplied by the combustion of fuel oil containing vanadium; and optionally sulfur and sodium, or other corrosive ingredients.

Although the present invention has been described in conjunction with preferred embodiments, it is to be understood that modifications and variations may be resorted to without departing from the spirit and scope of the invention as those skilled in the art will readily understand. Such modifications and variations are considered to be within the purview and scope of the invention and the appended claims.

What is claimed is:

1. A non-austenitic heat and corrosion resistant alloy adapted for use in hot vanadium-containing gaseous at-

mospheres consisting essentially by weight of about 0.8 to 1.2% carbon, about 1 to 2.2% silicon, about 0.4 to 10% manganese, about 23 to 35% chromium, about 2.5 to 11% tungsten, about 1 to 4% molybdenum, 0 to about 2% each of an alloying element selected from the group consisting of cobalt, nickel, niobium, tantalum, vanadium and titanium, 0 to about 1% aluminum, 0 to about 0.1% boron, 0 to about 1% nitrogen, 0 to about 4% copper, rhenium ranging from 0 to about 3.3% and 0 to about 3% total of misch metal, and the balance essentially iron, said alloy being characterized by the presence of ferrite and sigma phases.

2. The alloy of claim 1, in which the amount of essential elements range from about 0.8 to 1.2% carbon, about 1.8 to 2.2% silicon, about 0.4 to 10% manganese, about 28 to 35% chromium, about 7 to 11% tungsten, and about 1 to 4% molybdenum.

3. The alloy of claim 1, in which the amount of essential elements range from about 0.8 to 1% carbon, about 1.5 to 2% silicon, about 1 to 10% manganese, about 23 to 32% chromium, about 2.5 to 9% tungsten, and about 1 to 3% molybdenum.

4. As an article of manufacture, a heat and corrosion resistant structural element adapted for use in corrosive vanadium-containing environment at temperatures of over about 500°C, said element being formed of an alloy consisting essentially by weight of about 0.8 to 1.2% carbon, about 1 to 2.2% silicon, about 0.4 to 10%

manganese, about 23 to 35% chromium, about 2.5 to 11% tungsten, about 1 to 4% molybdenum, 0 to about 2% each of an alloying element selected from the group consisting of cobalt, nickel, niobium, tantalum, vanadium and titanium, 0 to about 1% aluminum, 0 to about 0.1% boron, 0 to about 1% nitrogen, 0 to about 4% copper, rhenium in amounts ranging from 0 to about 3.3% and 0 to about 3% total of misch metal, and the balance essentially iron.

5. The article of manufacture of claim 4, wherein the amount of essential elements ranges from about 0.8 to 1.2% carbon, about 1.8 to 2.2% silicon, about 0.4 to 10% manganese, about 28 to 35% chromium, about 7 to 11% tungsten, and about 1 to 4% molybdenum.

6. The article of manufacture of claim 4, wherein the amount of essential elements ranges from about 0.8 to 1% carbon, about 1.5 to 2% silicon, about 1 to 10% manganese, about 23 to 32% chromium, about 2.5 to 9% tungsten, and about 1 to 3% molybdenum.

7. The article of manufacture of claim 4, wherein said structural element is a valve seat formed of said alloy.

8. The article of manufacture of claim 5, wherein said structural element is a valve seat formed of said alloy.

9. The article of manufacture of claim 6, wherein the structural element is a valve seat formed of said alloy.

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